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The use of resonant X-ray magnetic scattering to examine the magnetic phases in UAs-USe solid solutions

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Abstract

Recent discoveries in the study of magnetism using synchrotron radiation are helping to elucidate the complex magnetism of the actinides. In this article we discuss the use of resonant X-ray magnetic scattering to investigate multi-**k** structures and the existence of induced magnetic moments in UAs-USe solid solutions. We present magnetic scattering experiments conducted at the ID20 and XMaS beamlines at the ESRF, Grenoble, France, and the ID-1 SRI-CAT beamline at the APS, Argonne, USA. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The uranium monopnictides and monochalcogenides with the fcc structure display a fascinating variety of magnetic structures. The uranium pnictides have an antiferromagnetic spin arrangement with 1**k**, 2**k** and 3**k** structures upon traversing the series, and all of the uranium chalcogenides are ferromagnets. Although these materials have been characterized by neutron techniques [1] the origin of the multi-**k** structures is not yet fully understood. Recent discoveries in the study of magnetism using synchrotron radiation are beginning to elucidate these complex magnetic materials. For example, resonant X-ray magnetic scattering (RXMS) has been used to examine the nature of the couplings leading to the 2**k** structure [2] of UAs, and has also revealed spin polarization of the As electronic states [2,3]. In this article we report experiments that examine the interplay of the RXMS cross section with multi-**k** structures and examine resonant scattering at the As and Se K edges of the UAs-USe solid solutions.

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2. Resonant scattering in UAs-USe solid solutions

The UAs-USe solid solutions are ideal for these experiments with their variety of multi-**k** structures [4]. At 12 K these materials have a 1**k**, 2**k** or 3**k** antiferromagnetic structures or order ferromagnetically, depending upon the As/Se content. For a type-IA $(++--)$ 1**k** structure, the Fourier components of the magnetization are directed along the propagation vector $\mathbf{k}_3=[0,0,1/2]$. The 2**k** structure is formed by the superposition of 2 orthogonal propagation vectors, for example $\mathbf{k}_1=[1/2,0,0]$ and $\mathbf{k}_2=[0,1/2,0]$, with the resultant moments in the $(0,0,1)$ plane. The 3**k** structure is made of 3 orthogonal propagation vectors and the resultant moments are orientated along the $\langle 1,1,1\rangle$ directions. We present magnetic scattering experiments conducted at (1) ID20 at the ESRF to study the multi-**k** resonant harmonics, (2) XMaS at the ESRF to examine the resonant scattering at the As and Se K edges, and (3) ID-1 SRI-CAT at the APS, USA, to observe resonant scattering from a ferromagnetic material.

3. Multi-k magnetic structures

3.1. The resonant harmonics of the X-ray magnetic scattering cross section

The RXMS amplitude for $E1$ (electronic dipole) transitions can be written [5]

$$f_{n,\mathrm{E1}}^{\mathrm{RXMS}}=[(\varepsilon'\cdot\varepsilon)F^{(0)}-i(\varepsilon'\times\varepsilon)\cdot\mathbf{z}_nF^{(1)} + (\varepsilon'\cdot\mathbf{z}_n)(\varepsilon\cdot\mathbf{z}_n)F^{(2)}],$$

where ε and ε' are the incident and scattered polarization vectors, and $\mathbf{z}_n$ is a unit vector in the direction of the magnetic moment on the nth atom. The factors $F^{(i)}$ are related to the matrix elements between the core and excited states and give the oscillator strength. The first term in $F^{(0)}$ contributes to the charge peak. The second term in $F^{(1)}$ is linear in $\mathbf{z}_n$ and gives the familiar strong dipole resonant amplitude for actinides, which appears at $\mathbf{Q}=\mathbf{G}+\mathbf{k}$, where $\mathbf{Q}$ is the momentum transfer, $\mathbf{G}$ is a Brillouin zone center, and $\mathbf{k}$ is the magnetic wave vector. If the incident polarization is σ then the final polarization is rotated to the π channel. The third term $F^{(2)}$ is quadratic in $\mathbf{z}_n$ and occurs at $\mathbf{Q}=\mathbf{G}+2\mathbf{k}$ or, with a multi-**k** structure, may occur at $\mathbf{Q}=\mathbf{G}+\mathbf{k}_1+\mathbf{k}_2$, where $\mathbf{k}_1$ and $\mathbf{k}_2$ are independent propagation vectors of the magnetic structure [2]. For incident σ polarization, the scattered polarization resulting from the $F^{(2)}$ term may be mixed. More surprisingly, in the study reported here, we find peaks at $\mathbf{Q}=\mathbf{G}+\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$ for a 3**k** structure. The origin of these weak peaks requires further consideration.

3.2. Multi-k resonant harmonics

The aim of this experiment was to search for a resonant harmonic associated with the 3**k** magnetic structure. The $x=0.2$ crystal is of particular interest for this study because there is evidence for the existence of both 2**k** and 3**k** magnetic structures [6], with the 3**k** phase dominating for $T>60$ K. The crystal was cooled to 70 K in the 3**k** phase. The photon energy was tuned close to the uranium M_4 edge and an Al $(1,1,1)$ crystal was used for polarization analysis. The $\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$ satellite observed for the first time is presented here. Fig. 1 shows the energy dependence of the $(0,0,2+k)$ and $(k,k,2+k)$ resonant harmonic satellites with $k=1/2$. The $\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$ satellite shows the same energy dependence as the first

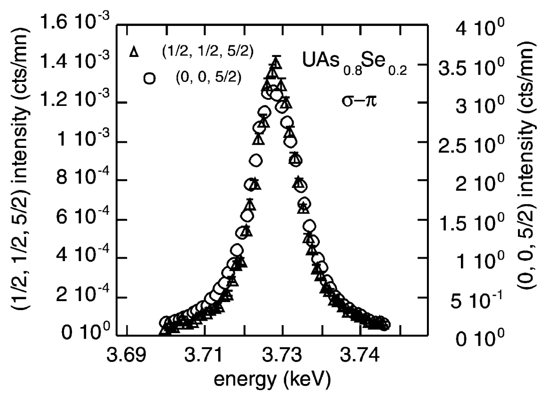

Fig. 1. The energy dependence of the first $(\mathbf{k}_3)$ resonant harmonic close to the U M_4 edge (3.728 keV). The energy dependence of the third $(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3)$ resonant harmonic close to the U M_4 edge (3.728 keV). Intensities are given an ordinate axis and are arranged to coincide at the peak.

resonant harmonic resonant satellite, $\mathbf{k}_3$, which is of dipolar origin. Resonant scattering was only observed in the $\sigma \rightarrow \pi$ polarization channel. Below the uranium M_4 edge no magnetic signal was observed at $(k, k, 2+k)$. A much reduced intensity for the $\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$ satellite was observed in the mainly $2\mathbf{k}$ phase below 50 K. The significance of these $\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$ resonant peaks is that they could be a signature of the $3\mathbf{k}$ magnetic structure. This is important since in standard diffraction experiments it is only possible to distinguish an assembly of $1\mathbf{k}$ magnetic domains from a multi-$\mathbf{k}$ structure by applying a symmetry-breaking magnetic/electrical/stress field, which would favor a particular domain. Further theoretical developments in reconsidering the resonant X-ray cross section are under way to account for these new observations.

4. Anion K edge resonance

4.1. Antiferromagnetic resonant scattering at the As and Se K edges

The discovery of unexpectedly large resonant enhanced scattering at the K edge of nominally non-magnetic anions [3] has stimulated the need to characterize this phenomenon. Our aim in this experiment was to characterize the K edge resonance and to relate the scattering intensity to the As/Se ratio. We present results for the $x=0.2$ crystal, which at 12 K has a $2\mathbf{k}$ antiferromagnetic structure. Polarization analysis was performed using an Al (4, 4, 0) crystal. Figs. 2(a) and 3 show the resonant scattering from the $(0, 0, 6\text{-}k)$ reflection, with $k=1/2$. Resonant scattering was observed in the $\sigma \rightarrow \pi$ polarization channel at both the As and Se K edges. The $x=0.1$ crystal was also studied and resonant scattering was again observed at the K edges of both anions. Analysis of these results has revealed that the intensity ratio of the peak resonance at the As and Se edges is related to the As/Se content.

4.2. Ferromagnetic resonant scattering at the As K edge

Our aim was to confirm that we could observe resonantly enhanced scattering at the K-edge of a non-magnetic anion in a ferromagnetic material. For this experiment the $x=0.5$ composition, which is a ferromagnet below 165 K, was used. A permanent magnet of 0.4 T was used to induce a preferential domain orientation along the moment direction before cooling the crystal to 100 K. Circular polarization was obtained from a diamond quarter-wave plate. The photon helicity was switched

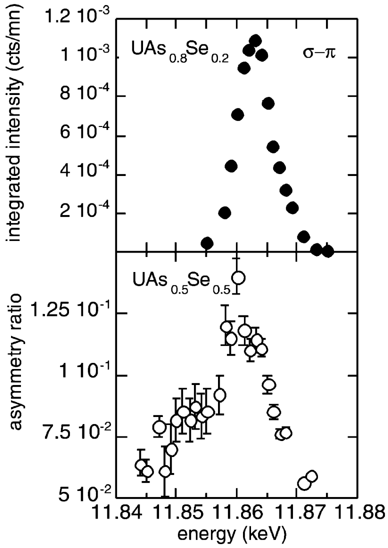

Fig. 2. (a) Resonant scattering in the $\sigma \rightarrow \pi$ polarization channel close to the As K edge (11.867 keV) of antiferromagnetic UAs-USe. (b) Asymmetry ratio as a function of energy close to the As K edge (11.867 keV) of ferromagnetic UAs-USe.

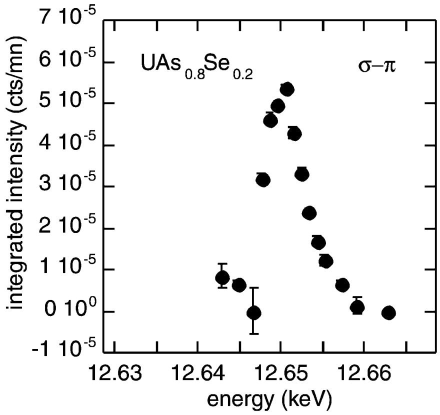

Fig. 3. Scattering in the $\sigma \rightarrow \pi$ polarization channel close to the Se K edge (12.658 keV).

to reverse the interference between the charge and magnetic scattering. The energy dependence of this asymmetry ratio is shown in Fig. 2(b). This preliminary result suggests that this ferromagnetic material exhibits resonantly enhanced scattering at the As K edge.

5. Conclusion

The technique of resonant magnetic scattering is still developing and opening up new fields of study. Firstly, with the discovery of $\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$ resonant magnetic satellites in the 3**k** phase of the UAs-USe solid solutions, and with further theoretical developments, it may be possible with resonant X-ray magnetic scattering to distinguish between different multi-**k** structures without having to apply a symmetry-breaking field. Secondly, the observation of unexpectedly large enhancements at the K edges of the anions in uranium ferro and antiferromagnets opens up further possibilities for diffraction experiments as an element-specific probe, which are not restricted to the relatively low energy of the uranium M edges. These results not only suggest new fields in the study of actinide materials, but also illustrate the need to further examine the basic formalism of the resonant scattering process.

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